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A Qualitative Analysis of the Neutron Population in Fresh and Spent Fuel Assemblies during Simulated Interrogation using the Differential Die-Away Technique

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Abstract:

Monte Carlo simulations were performed for the differential die-away (DDA) technique to analyse the time-dependent behaviour of the neutron population in fresh and spent nuclear fuel assemblies as part of the Next Generation Safeguards Initiative Spent Fuel (NGSI-SF) Project. Simulations were performed to investigate both a possibly portable as well as a permanent DDA instrument. Taking advantage of a custom made modification to the MCNPX code, the variation in the neutron population, simultaneously in time and space, was examined. The motivation for this research was to improve the design of the DDA instrument, as it is being considered for possible deployment at the Central Storage of Spent Nuclear Fuel and Encapsulation Plant in Sweden (Clab), as well as to assist in the interpretation of the both simulated and measured signals.

Keywords: differential die-away, spent fuel, used fuel, active neutron interrogation

1. Introduction

The differential die-away (DDA) technique [1] is one of several nondestructive assay techniques investigated as part of the Next Generation Safeguards Initiative Spent Fuel (NGSI-SF) Project [2,3]. The purpose of the NGSI-SF project is to use nondestructive assay (NDA) technologies to strengthen the technical toolkit of safeguard inspectors and others to determine the following technical goals more easily and more accurately:

- Detect replaced or missing pins from spent fuel assemblies (SFA) to confirm item integrity and deter diversion,
- Determine plutonium mass and related plutonium and uranium fissile mass parameters in SFAs, and
- Verify initial enrichment, burnup, and cooling time of facility declaration for SFAs.

The project also includes two related goals with facility-specific benefits: (1) estimation of the heat content and (2) estimation of reactivity (multiplication).

The DDA technique as deployed involves the active interrogation of a fuel assembly with a burst of neutrons. The signature typically used in analysis comes from integrating the counts in a time interval within the dynamically evolving neutron population in the nuclear fuel assembly. The DDA technique can be used to measure the multiplication of a fuel assembly [6], to estimate the plutonium mass [7] or the content of fissile material in the form of the effective ^{239}Pu mass ($^{239}Pu_{eff}$) [8], to estimate the initial enrichment, burnup, and cooling time of the fuel [9] and to detect missing fuel, however, these capabilities are not the primary subject of this paper. Ideally it will be possible to make such determination without using operator declaration; it is part of the NGSI-SF Project research plan to

examine the capability of individual nondestructive assay (NDA) techniques as part of the overall NGSI-SF Project research plan of assessing integrated NDA systems in pursuit of the identified goals.

The DDA technique actively interrogates a fuel assembly leading to the release of primarily prompt neutrons from the fission of fissile, and to a lesser degree fertile, material in the spent fuel assembly. A variety of pulsed neutron source can be used for this active interrogation; in the current investigation a deuterium-tritium (DT) neutron generator was selected. As the system is subcritical, the induced neutron population decreases with time with a half-life that is on the order of hundreds of microseconds. The measured DDA signal reveals various properties of the fuel assembly, such as multiplication, which can be used to characterize the fuel assembly. Additionally the DDA signal was previously shown to be a function of the isotopic mixture that results from the initial enrichment, burnup, and cooling time of the fuel. For this current research the MCNPX code [4,5] was used to simulate the time and spatial variation of the neutron population for three different DDA setups.

The presented simulations are unique relative to past research in that the 3-dimentional spatial evolution of the neutron population is displayed in 2-dimensional images in time. This unique description of DDA performance was conducted with both fresh and spent fuel assemblies. The primary goal of this current work is to examine the DDA signal in both a spatial and temporal way in order to suggest possible design changes that may better achieve the technical goals of the NGSI-SF Project, particularly in the context of encapsulation/repository safeguards. Previous DDA simulations, which produced favourable results, [6] used a large mass of metal for spectrum tailoring of the generator neutrons and moderation. The original instrument design [10] used ^{3}He neutron detectors, which in a spent fuel environment, required lead (Pb) shielding in order to protect the ^{3}He tubes from gamma radiation. Additionally the previous work focused only on ^{3}He detectors, while this current work, researched a lightweight design of the DDA instrument, included fission chambers which are less sensitive to gamma radiation, making the thick Pb shielding unnecessary. Fission chambers have approximately 1% the detection efficiency per unit length as compared to ^{3}He tubes for thermal neutrons. Since this inefficiency impacts both the signal and the background equally, and the counting rates are high, it is not expected to impact the DDA instrument performance significantly.

2. DDA instrument design

Four different DDA instrument designs are discussed in this paper. Each is described below and illustrated in Fig. 1 A through D. Common to all the designs is the use of 8 tubes, either ^{3}He or fission chambers. Also in each design, the detector tube is surrounded by a few cm of polyethylene that is then surrounded by Cd. The polyethylene is included to moderate, and as such, to increase the detection probability of neutrons that were high enough in energy to penetrate the Cd layer. The Cd layer absorbs essentially all neutrons with energies lower than 0.46 eV and is needed in order to ensure that the DDA detectors detect non-thermal neutrons only. In all cases the assemblies and the DDA instrument are submerged in fresh water. Furthermore, all DDA simulations track only the neutrons produced by the neutron generator during a 10 μs burst as well as the neutrons released in fission chains initiated by these neutrons. The neutrons from fission chains started by neutrons released in spontaneous fission in the fuel are not simulated

The design illustrated in **Fig. 1A** [10] represents the NGSI-SF Project design at the start of this current research. A key driver in the design depicted in Fig. 1A was the need at that time to integrate DDA with delayed neutron detection. The inclusion of delayed neutron detection drove two key features of the design: (1) thick spectrum tailoring material was included to reduce the neutron energy in order to reduce the fission of ^{238}U because ^{238}U is more than 80% of the fuel by weight and ^{238}U is a strong delayed neutron source per fission (~7 times stronger than ^{239}Pu per fission). (2) As labelled in Fig. 1A, only 6 of the ^{3}He tubes were used to generate the DDA signal because only those tubes were covered in cadmium. The two tubes not surrounded in cadmium have elevated detection efficiency and were used only for delayed neutron detection. The ^{3}He tubes had a 1.89 cm diameter and a 5.08 cm length. The instrument simulated was made to fit a 17 x 17 assembly with a 5 mm water gap between the assembly and the detector walls on all sides.

The design illustrated in **Fig. 1B** represents the first iteration towards the lightweight version of the DDA instrument envisioned for a portable use and varies relative to Fig. 1A in the following ways: The square high density polyethylene blocks were replaced with cylinders for which each cylinder is

individually lined with cadmium. The metal spectrum tailoring was replaced with water as a “DDA only” instrument can use a weaker neutron generator and function with a higher energy neutron interrogating spectrum. The intended application of Fig. 1B was fresh fuel measurements for the purpose of simulation validation. Lead shielding was maintained between the fuel and the detector even though it was not needed for gamma attenuation in order to reduce the number of variables changed between the two designs and as a starting point for assessing the impact of the lead. The instrument simulated was made to fit a 15 x 15 assembly with a 5 mm water gap between the assembly and the detector walls.

The design depicted in **Fig. 1C** is very similar to that of Fig. 1B with the exception that the lead shielding was eliminated which allowed the ^{3}He detectors to be moved closer to the fuel. The instrument simulated was also made to fit a 15 x 15 assembly but the water gap between the assembly and the detector walls was doubled to 10 mm.

The design depicted in **Fig. 1D** is modified from that illustrated in Fig. 1C in one significant way. A 1 mm layer of cadmium was added between the neutron generator and the fuel. The water gap was also reduced slightly to allow for a 17 x 17 assembly to be used and fission chambers were simulated instead of ^{3}He. This later change lengthens the die-away time of the individual detectors. The fission chamber tubes had a 2.4 cm diameter, and 5 cm length.

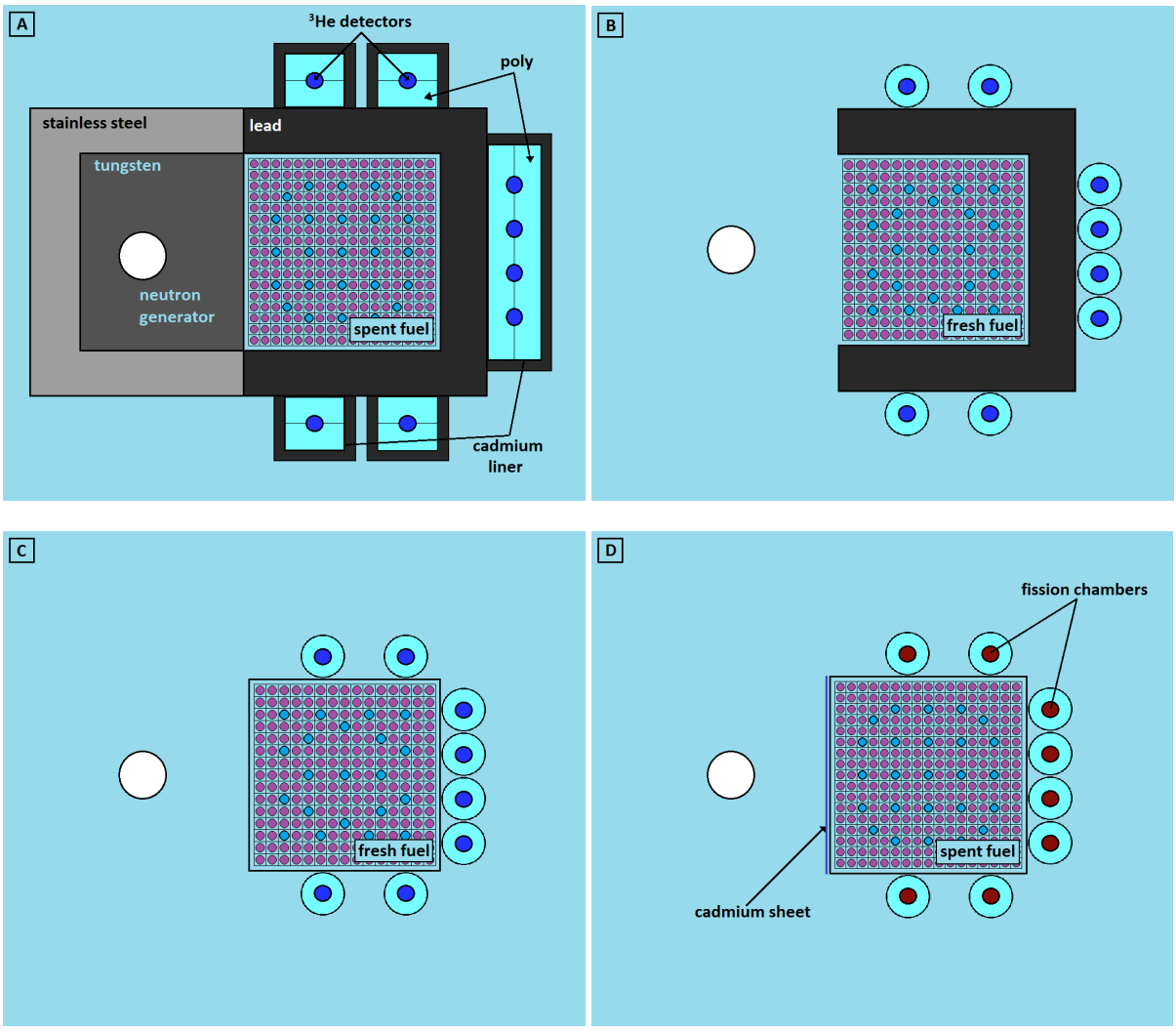

Figure 1. Schematic cross sectional view of the four DDA instrument designs investigated in this study: (A) the spent fuel design used by the NGSI-SF Project at the start of this current research effort, (B) the fresh fuel design that uses water for spectrum tailoring and has individual cadmium lined tubes, (C) the same design as (B) but without the lead shielding which allowed the tubes to move closer to

the fuel, and (D) the spent fuel design that is essentially the same as design (C) with the addition of a cadmium sheet and the ^{3}He tubes were replaced by fission chambers.

3. The fuel simulated

A range of pressurized water reactor fuel, both fresh and spent, was simulated in the course of this research. Yet, in this study the results for only one fresh and one spent fuel case are depicted. The fresh case was for a 15x15 assembly comprised of ^{235}U to an enrichment of 3.19 wt-% with 204 fresh fuel rods (0.475 cm diameter and 131 cm length) and 21 water-filled guide tubes. This specific case was selected to match planned experiments at Los Alamos National Laboratory (LANL). A "fully burnt" assembly was selected for the SFA simulations because the overwhelming majority of spent fuel in the world is fully burnt. Using the simulation capability in the MCNPX code, a Westinghouse 17x17 with an initial enrichment of 4 wt-% was irradiated to have a burnup level of 45 GWd/tU and then cooled for 5-years; this fuel was part of the NGSI-SF Library 1 [11]. The purpose of simulating fresh fuel in a project that is primarily interested in spent fuel is to make sure that we can benchmark our simulations in an environment that is well characterized.

4. Temporal description of neutron population in fresh fuel

The DDA system simulations were performed using a customized version of MCNPX v273[4,5]. In Fig. 2A and 2B, the number of detected neutrons per neutron generator source neutron for the design depicted in Fig. 1B and Fig. 1C, respectively, are illustrated as a function of time. The "first fission" tallying capability was used to identify if the detected neutron (1) originated in the neutron generator, (2) was part of a chain of events for which the first fission in the chain was ^{238}U, (3) was part of a chain of events for which the first fission in the chain was ^{235}U. The MCNPX capability to identify a detected neutron according to the first induced fission in its chain of interaction was added to the MCNPX code explicitly for this work and is called the "first fission" capability. With the first fission capability, the detected fission contribution from individual isotopes (uranium isotopes for fresh fuel and also plutonium isotopes for spent fuel), and the detected neutrons from the DT neutron generator pulse were tallied to indicate their origin. In these simulations all neutrons start in the neutron generator. If, as a neutron advances through the simulation, it should cause a fission, the identifier or tag on that neutron is changed from originating in the neutron generator to originating in the isotope that underwent fission. From that time onward, the neutron maintains the label it received on its first fission. Analysing these values provides meaningful information on the interaction of the interrogating pulse with the fuel. Additional mesh tallies in the simulation were used to plot the dynamic evolution of the neutron spatial distribution in time for a visual and qualitative understanding of its behaviour.

Basic characteristics of DDA, instrument design 1B and 1C, as applied to spent fuel assemblies are evident in both Fig. 2A and 2B; the detected burst neutrons peak in the time domain before 20 μs. These neutron generator originating neutrons quickly decrease and within 200 μs they are about two orders of magnitude less than the neutrons that originate from ^{235}U actively induced fission. After 50 μs, the neutrons that originate from ^{235}U actively induced fission begin to dominate the detected neutron signal for both the case with the large lead shielding and without it.

There are some differences evident between Fig. 2A and 2B. The detected neutrons that first fission in ^{235}U and ^{238}U both increase in the progression from the design depicted in Fig. 1B to that of 1C; the percentage increase was greater for ^{238}U. The detection efficiency for neutrons created in the assembly increased as we move from the Fig. 1B design to the Fig. 1C design because the ^{3}He detectors moved closer to the fuel and the water layer around the fuel doubled, which increased the multiplication of the assembly. The main point for studying the geometries depicted in Fig. 1B and Fig. 1C was to learn if the illustrated change in geometry had an impact on the ability of the DDA instrument to discern among a range of spent fuel assemblies. It was observed that the temporal variation of the detected neutron signal was a consistent trend with spent fuel variation between the two designs. Hence, leading to the conclusion that both designs are viable designs.

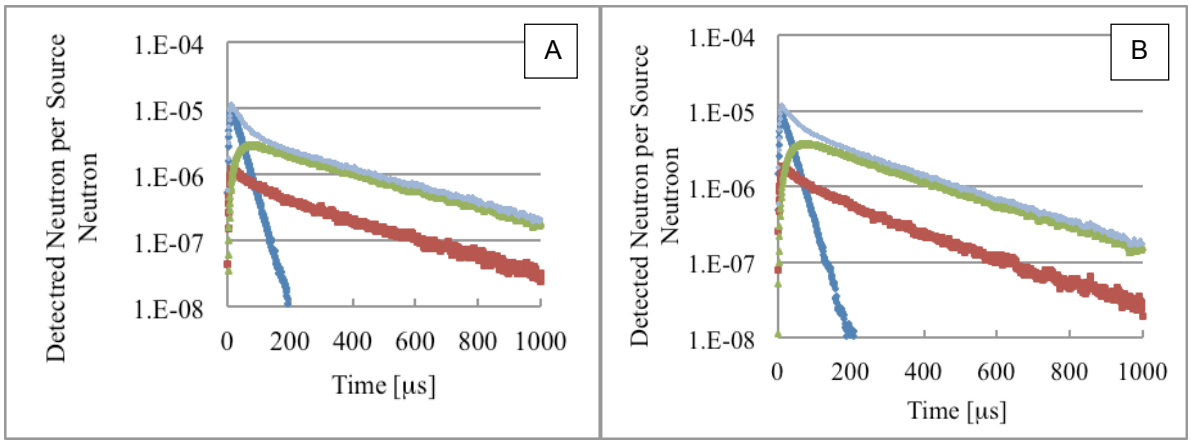

Figure 2. The number of detected neutrons per source neutron for the physical setup illustrated in Fig. 1B and Fig. 1C are depicted for A and B, respectively. The "first fission" tallying capability was used to identify the detected neutrons as originating in the neutron generator (dark blue), ^{238}U (red) or ^{235}U (green); the sum of all detected neutrons are indicated in light blue.

5. Spatial description of neutron population in fresh fuel

In order to understand the spatial qualities of the neutron die-away, the MCNPX code was altered to record a two dimensional depiction of the neutron population as a function of time. This graphical depiction, known in MCNPX terminology as a "mesh tally," was made time-dependent specifically for this NGSI-SF DDA research effort. For the results of this research the neutron population was integrated over voxels that were 7.0 mm by 5.6 mm in the horizontal plane and 310 mm in the vertical plane. In Fig. 3A and 3B, the relative neutron population averaged over two separate time intervals, 10-30 µs and 190-210 µs respectively, for the DDA design illustrated in Fig. 1C are illustrated. The first time interval was selected because that is the interval of time just after the neutron generator is shut off. The later time interval was selected to be consistent with spent fuel simulations illustrated in the next section. Note that the colour scales are not consistent between the two images; yet in both images there is essentially a factor of 3 variation between the pin in which the neutron population is greatest and the pin for which it is least. There is only a slight change in the relative spatial distribution of the neutron population between these two images, which is consistent with the data observed when the spatial images are viewed as a movie. In other words, the neutron population starts and stays relatively centred in the assembly during the measurement duration. From 10 to 70 µs there is only a slight motion of the distribution from the neutron generator side of the assembly toward the centre; from 70 µs out to 470 µs the neutron population remained nearly centred in the assembly. The ending time of 470 µs was chosen because the simulation statistics became poor. Because the neutrons originating in the neutron generator are such a significant part of the detected neutrons signal in these early times, before ~50 ms, the traditional DDA signal, which does not include the interrogating source, can be obtained after ~50 ms when the neutron population is centred in the assembly.

It is important to emphasize that the images in Fig. 3 are for the neutron population. The detected signal can be connected to the neutron population by the detection probability per pin. This is outside the scope of this research. This connection is assembly specific since it depends on the multiplication of the assembly. For high multiplying assemblies neutron starting in the centre have a greater chance of starting a chain reaction that results in a detected count than do neutrons originating from the centre of a low multiplying assemblies. Although, results for specific assemblies are not presented here, research done with other neutron instruments in the NGSI-SF project have illustrated that in general the detection probability is elevated for neutron originating in the edge of the assemblies particularly for fully burnt assemblies that make up the vast majority of commercial spent fuel assemblies. Hence the combination of the peaked neutron population illustrated in Fig. 3 with the oppositely shaped detection probability distribution is anticipated to make the spatial origins of the detected signal more uniform than the distributions depicted in Fig. 3; yet, the degree to which this "balancing" occurs will be assembly dependent and will be a topic of future research.

The location and density of the neutron population is driven by multiplication, which depends on the fuel composition, as well as the material surrounding the assayed fuel assembly. The population decreases more slowly in regions where the multiplication is the greatest. It is important to note that the fresh fuel case depicted in Fig. 3 is among the more highly multiplying cases expected. This case can be thought of as a bound in terms of the multiplication to be expected from an assembly. The 15 x 15 assembly simulated in Fig. 1C contained rods that were 3.19 wt-% ^{235}U resulting in a net multiplication of ~3.1 for Watt fission spectrum neutrons emitted uniformly from within the assembly with the assembly isolated in fresh water. A typical commercial "fully burnt" 17 x17 assembly, would have a net multiplication of approximately ~2.1, while "fully burnt" smaller assemblies would have still lower multiplication values. This case is presented here for three reasons: (1) it is a bounding case given its high multiplication being similar to a one cycle irradiated assembly that started with ~4.2% ^{235}U initial enrichment, (2) it was anticipated to be one of the measurement setups used at LANL and (3) it provides context for the interpretation of spent fuel results presented later in this study.

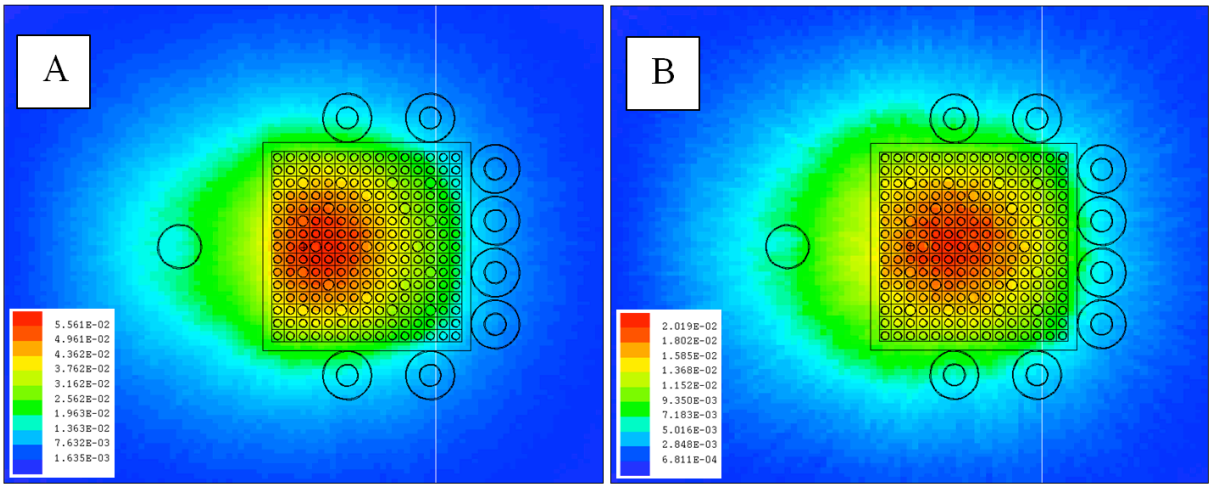

Figure 3. Spatially relative neutron population integrated over 20 μs intervals for the fresh fuel case and DDA design illustrated in Fig. 1C for the two time intervals: (A) 10-30 μs, (B) 190-210 μs (Note: units are in neutrons per source particle over a time interval, but the colour scales are not consistent between the two plots.).

6 Spent fuel simulation results

The simulation of fresh fuel measurements was the focus of this research to this point because the research effort is preparing for fresh fuel benchmarking measurements. Yet, because the ultimate goal of the NGSI-SF Project is spent fuel, in this section we present several simulations of spent nuclear fuel assay with the lightweight DDA design. In Fig. 4 A to D the relative neutron population integrated over four different 20 ms intervals is illustrated for a 4% wt. initial enrichment, 45 GWd/tU burnup, 5-year cooled assembly; this assembly was chosen since it has roughly the same multiplication as the vast majority of "fully burnt" 17x17 assemblies. The DDA design simulated is the one illustrated in Fig. 1C however with a 17x17 spent fuel assembly. In changing from the 15x15 fresh fuel design used for Fig. 1 and 2 to the 17x17 spent fuel case the assembly width was 1.6 mm larger than the width of the 15x15 assembly, so it was decided to leave the structure of the instrument the same and to allow the water gap around the assembly to be reduced slightly to accommodate this slightly larger assembly.

In contrast to the fresh fuel simulations, the spatial peak in the neutron population in Fig. 4 exhibits a more dynamic behaviour. It appears to migrate into and then out of the spent fuel assembly. The "apparent movement" of the neutron population indicates that the neutron population lives longer in certain regions of the assembly than it does in the other regions. In particular the neutrons live longer in the boundary region at the edge of the assembly (i.e. in the water/fuel boundary) on the neutron generator side of the fuel as compared to the neutron population in either the centre of the assembly or in the boundary region between the fuel and the Cd lined detectors. The population changes among these three regions because the combined effects of the materials in each region foster future generations more or less. The boundary region on the neutron generator side of the assembly produces a larger multiplication in that region as compared to the region in the centre of the assembly

or the boundary region on the side opposite to the neutron generator (NG). The boundary region on the three sides of the SFA is especially impacted by the presence of Cd around the detector tubes, which reduces the multiplication in these water/fuel interface areas relative to the water filled neutron generator side of the assembly. In other words, the presence of a relatively thick water region on the left side of the assembly depicted in Fig. 4, for which the absorption cross section is low, elevates the multiplication in the left edge region of the assembly.

The multiplication of the fuel itself changed from 3.1 to 2.1 between the fresh fuel case depicted in Fig. 3 and this spent fuel case depicted in Fig. 4. With the multiplication change the neutron population went from one that was nearly stationary in time and space to a population that varies in time and space. It is important to note that the difference in isotopic composition between the two cases is vast with the fresh fuel case only containing ^{235}U, ^{238}U and ^{16}O while the spent fuel case has a large mixture of actinides and fission products each with its own unique cross sections (absorption and/or fission). The composition of the fuel also changed from the uniform pin-to-pin case of fresh fuel to the non-uniform case of an irradiated assembly. The non-uniformity, in this particular case, is the same in all directions outward from the centre of the assembly because in the calculation of the burn-up of the assembly the model assembly had infinitely reflecting boundary conditions. Hence the non-uniformity of the pin-to-pin variation does not explain why the neutron population appeared to move from near the centre to the left.

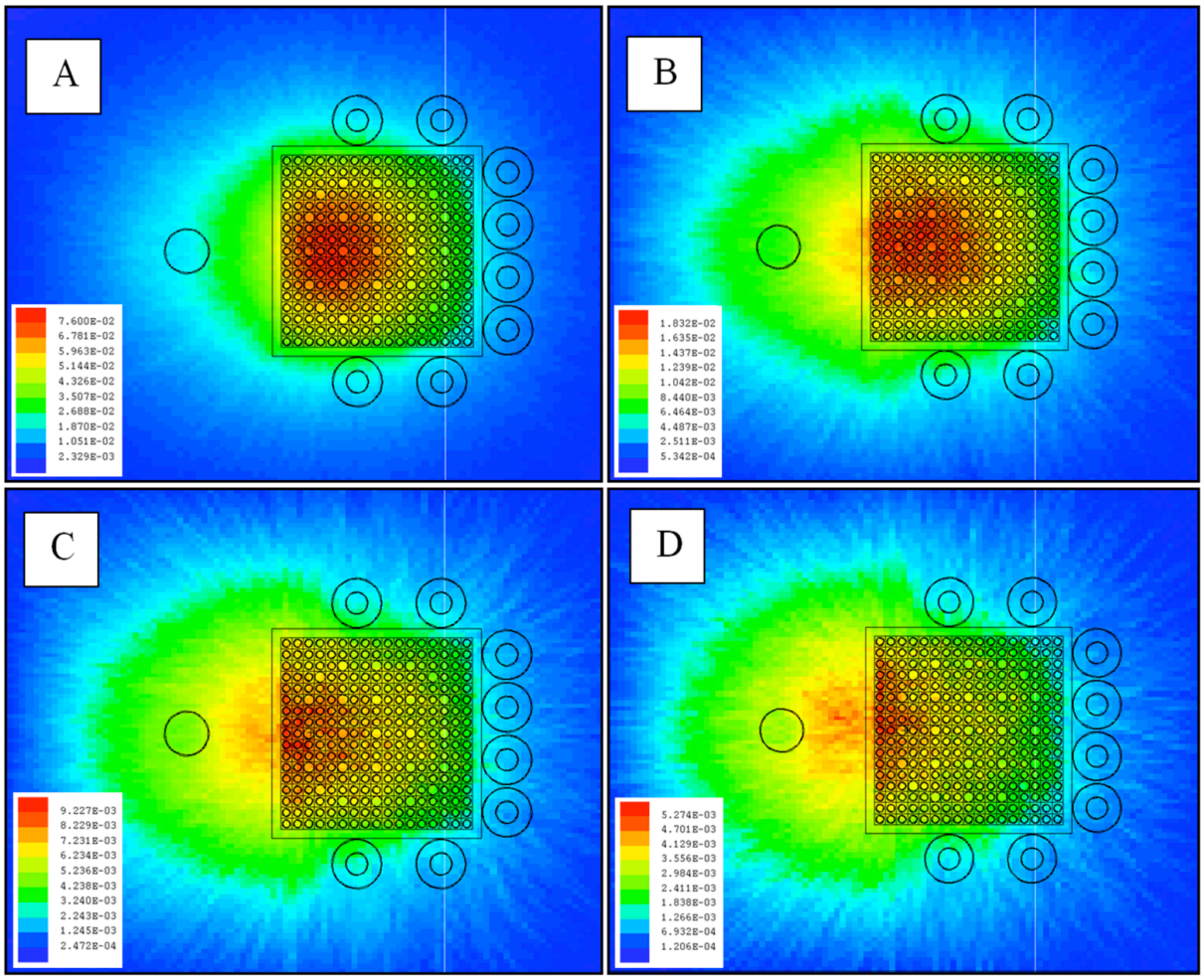

Figure 4. Spatially relative neutron population integrated over 20 μs intervals for the spent fuel case and DDA design illustrated in Fig. 1C for time intervals (A) 10-30 μs, (B) 110-130 μs, (C) 190-210 μs, and (D) 290-310 μs (Note: The color scales are not consistent between the four plots.)

From the data illustrated in Fig. 3 and Fig. 4, cases for which the only major change was the fuel itself, we can conclude that the location of the peak in the neutron population in an assembly depends on the assembly itself. The two cases examined illustrate that even in the same environment around the

fuel assembly the apparent evolution of the neutron population may be dramatically different both in time as well as space. The neutron population density in the 20 μs interval after the neutron generator was turned off is about the same in fresh and spent fuel (Figs. 3A and 4A, respectively). However, during the later time domain of 190-210 μs, the density of neutron population drops significantly in the spent fuel, while in the fresh fuel the decrease in the centre of the fuel assembly is only modest. The neutron population dies away much faster in the spent fuel than it does in the fresh fuel because of the reduced multiplication. Additionally, the neutron population on the NG side of the assembly is even larger in the fresh fuel case than in the same region of the spent fuel case. Yet in relative terms, this subpopulation is smaller in the fresh fuel case, and almost dominating in the spent fuel case. As freely as the neutrons can move from the fuel assembly into the water, they can also drift freely back into the fuel assembly. The probability of such re-entry per neutron is identical in both cases studied. While this influx of the thermal neutrons appears to be less significant in the case of fresh fuel, where a large neutron population still survives in the middle of the fuel assembly, it causes a large effect in case of the spent fuel.

From a different perspective, the thermal neutrons that re-enter the fuel from water act as an additional interrogating neutron source. But unlike the precisely controlled neutron generator, the overall intensity and time profile of this additional "neutron source" is not known, i.e. un-normalizable, and depends on the properties of the fuel assembly to be assayed and the boundary material. Moreover, should the SFA not have a constant burnup across all pins, this additional interrogation of the border region by the NG side could result in more or less severe oversampling of part of the SFA that may not be representative of its global characteristics. While this additional "interrogating capacity" can be relatively marginal and thus tolerated (fresh fuel), its effects may be overwhelming in other instances (spent fuel) and should thus be suppressed.

The solution for suppression of the return of the thermal neutrons is evident from the very pictures where this effect can be observed, (i.e. Fig.4). While the neutron population external to the SFA exists on the side of the NG, it is almost entirely absent on the other three sides of the instrument. This is not surprising given the extremely high absorption cross section of cadmium that lines each of the detectors present in these regions, and which effectively prevents any neutron subpopulation build-up. It is therefore suggested that to prevent neutrons thermalized in the water by the NG side of the SFA from re-entering the assembly, a thin Cd sheet be installed along the SFA side. Such cadmium sheet will absorb virtually all neutrons below 0.46 eV, hence virtually all thermal neutrons are prevented from returning to the fuel after scattering in the water.

As a proof of concept on the impact of cadmium, a simple modification was made to the cases simulated in Fig. 4. A 1 mm thick sheet of cadmium was added 10 mm away from the fuel between the neutron generator and the fuel as illustrated in Fig. 1D. This sheet was square and covered the entire side of the assembly. The neutron population for the same 4 time intervals depicted in Fig. 4 is illustrated in Fig. 5 for the same spent fuel assembly.

It is noteworthy that Fig. 4A and Fig. 5A, for the 10 to 30 μs time interval, are nearly identical in terms of the neutron spatial distribution. This is pointed out because the neutrons evolve very differently after this interval in time for the two cases. In the case with the cadmium liner the neutron population moves away slightly from the generator into a central position in the assembly and then stays there. Unfortunately, the neutron population distribution in the fuel in Fig. 5D is not clear because the neutron population located near the generator is so strong. It is worth pointing out that the neutrons outside the Cd liner depicted most prominently in Fig. 5D cannot be detected and cannot re-enter the fuel. The additional sheet of Cd, together with the combined effects of the Cd liners around individual fission chambers effectively isolated the SFA from its surrounding, thus preventing the secondary and undesired interrogation by neutrons reflected back into the fuel. The neutron population inside the fuel better reflects the properties of the fuel itself and spatially stabilizes at the place of the highest multiplication, which in Fig. 5 corresponds to the centre of the SFA.

An additional noteworthy comparison between Figures 4 and 5 involved comparing the neutron population in the 290-310 μs time interval in each figure. In Figures 4 the intensity of the maximum neutron population during this 20 μs time interval is difference in both intensity and location relative to Figure 5. The intensity of the maximum population when there is no Cd liner around the fuel, as in Figure 4, is double that which exists when the Cd liner is present, as in Figures 5; indicating that the over all neutron population is reduced by the presence of the Cd liner. Furthermore, when the Cd liner

is not present, the maximum population is located on the water-to-fuel interface indicating that this is the optimal region for the neutron population to endure in time. However, when a neutron liner passes through this water-to-fuel interface, the neutron population is severely reduced as indicated in Figures 5; from the colour scale in each figures a factor of 3 reduction of the neutron population, from around 5 x 10^{-3} to around 1.5 x 10^{-3} neutrons per source particle.

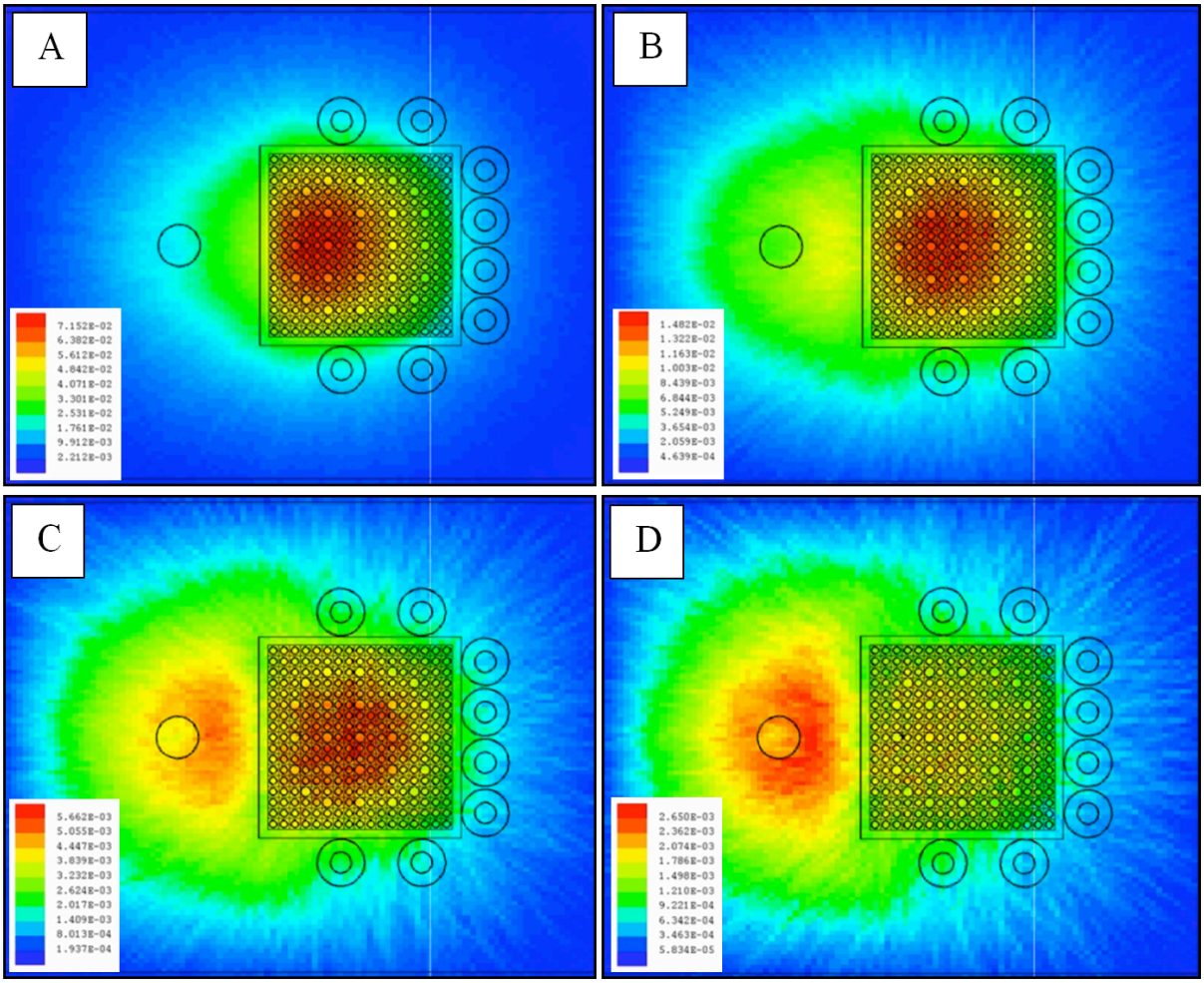

Figure 5. Spatially relative neutron population integrated over 20 μs intervals for the spent fuel case and DDA design illustrated in Fig. 1D for time intervals (A) 10-30 μs, (B) 110-130 μs, (C) 190-210 μs, and (D) 290-310 μs. The presence of Cd in the graph is not visible as it is only 1 mm thick. (Note: The colour scales are not consistent between the four plots.)

To further give context to this discussion, it is worth pointing out that a modification in the understanding of how DDA functions is suggested by the 2D images presented in this study especially together with the recent results on the DDA response to assay of asymmetrically burned SFAs [12]. Along with the delayed neutron constraint previously mentioned, the DDA design illustrated in Fig. 1A was designed with a mental expectation that there would be a fission gradient across the assembly at all times during the assay; the expectation was that the neutron source of the generator would make the neutron population highest on the neutron generator side of the assembly throughout the assay. The logic was that this elevated population near the generator results in elevated detection rates in nearby detectors and thus should be balanced by an oppositely varying efficiency gradient; hence, the detection efficiency should be greatest on the side far from the neutron generator and less near the neutron generator. Separate research has however indicated that different count rates each detector sees may be influenced by local properties of the SFA in its vicinity and that using a sum of all detectors can be inappropriate for characterization of average properties of the SFA [12]. Additionally the thermal return of neutrons from the water or other materials also influences detectors individually as it may cause the neutron population in the SFA to move at a time scale relative to the measurements. That motion can be influenced by both the material composition of the SFA as well as the material surrounding it. Therefore a couple design changes are suggested so that the behaviour of

the neutron population provide information about the assembly composition rather than the material around it: (1) a Cd layer around the entire assembly can isolate the assembly from its surroundings if necessary, and (2) detectors should be placed in multiple locations on three sides of the assembly to provide as complete of coverage as possible.

7. Conclusions

In this research three new DDA designs were compared to the NGSI-SF Project design that existed at the start of the research effort. The primary objective of this research is improving the design of the DDA instrument for possible deployment at the Swedish encapsulation facility. A new capability to visualize how the neutron population changes in space and time was utilized and has altered the conceptual understanding of how the neutron population evolves in the assembly. The existence or absence of a large lead layer around the assembly was determined to not significantly impact the performance of DDA for the cases studied. Additionally, one highly multiplying and one multiplying at the level of a fully burnt 17x17 assembly were simulated. The neutron population in both assemblies started in the 10 to 30 μs interval at a location a few rows off-centre. Given this as a starting point, it was shown that the centre of the neutron population can move in time. For the highly multiplying case it stayed nearly centred and for the lower multiplying case it moved to the edge of the assembly near the neutron generator apparently caused by the thermal neutrons returning from water into the fuel. Then it was illustrated that for this later case, the fuel assembly can be effectively isolated from its surrounding environment resulting in the neutron population reflecting the fuel composition rather than coupling it to its environment. The population could be manipulated to stay near the SFA centre by changing the edge conditions with Cd on that side of the assembly to which it had previously moved. The results presented here show that the location of the neutron population inside the assembly can essentially be independent of its surroundings for time intervals after ~50 ms. The alteration of the local multiplication in the edge changes the distribution of the neutron population within the assembly. Information learned about the neutron populations in the fresh and spent fuel assemblies from these simulations will be used to improve the designs of DDA instruments deployed in the near future, although varying circumstances merit different designs. The results presented here suggest that the application of a Cd liner is preferable when there is a significant amount of neutron reflecting material present, such as water, directly by or around the fuel assembly. Overall, the work benefitted the NGSI-SF project by understanding better the design challenges of an instrument that can determine the amount of fissile material present in a SFA, help detect partial defects and/or determine the amount of plutonium in the assembly, and to verify properties inherent to the assembly (i.e. initial enrichment, burnup, and cooling time).

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9. References

1. Caldwell W.E. and Kunz, J. T.; **"**Experimental Evaluation of the Differential Die-Away Pulsed-Neutron Technique for the Fissile Assay of Hot Irradiated Fuel Waste," Richland, Washington, ANS Topical Meeting on Treatment and Handling of Radioactive Waste, 1982.
2. Humphrey, M.A.; Tobin, S.T., and Veal, K.D.; "The Next Generation Safeguards Initiative's Spent Fuel Nondestructive Assay Project," Journal of Nuclear Material Management, Vol. 40, **3**:XL (2012)
3. Tobin, S.J., Menlove, H.O., Swinhoe M.T. and Schear, M.A.; "Next Generation Safeguards Initiative Research to Determine the Pu Mass in Spent Fuel Assemblies: Purpose, Approach, Constraints, Implementation, and Calibration," Nuclear Instruments and Methods in Physics Research A, vol. 652, no. 1, pp. 73-75, 2011.
4. Hendricks, J.S., McKinney, G.W., Wilcox, T.A., James, M.R.; “MCNPX 2.7.3 Extensions,” Los Alamos National Laboratory Report LA-UR-12-00135 (June 2012).

5. Hendricks, J.S. and Tobin, S.J.; “NGSI MCNPX Extensions to MCNPX 270,” Los Alamos National Laboratory Report LA-UR-12-00133 (January 2012)
6. Henzl, V., Swinhoe M.T., Tobin, S.J., Menlove, H.O.; "Measurement of the Multiplication of a Spent Fuel Assembly with the Differential Die-Away Method within the Scope of the Next Generation Safeguards Initiative Spent Fuel Project," Journal of Nuclear Material Management **3:**XL,p 61-69, (2012).
7. Henzl, V., Croft, S., Swinhoe, M.T., Tobin, S.J.; "Determination of Pu Content in a Spent Fuel Assembly by Measuring Passive Total Neutron Count Rate and Multiplication with the Differential Die-Away Instrument," Orlando, FL: Proceedings of Institute of Nuclear Material Management, 2012.
8. Lee, T.H., Tobin, S.J., Menlove, H.O., Swinhoe, M.T.; "Determining the Pu Mass in LEU Spent Fuel Assemblies: Focus on Differential Die-Away Technique," Los Alamos National Laboratory report LA-UR-11-00747 (2011).
9. Henzl, V., Swinhoe, M.T., and Tobin S.J.; "Determination of Initial Enrichment, Burnup, Cooling time of Spent Fuel Assemblies with a Differential Die-Away Techniques Based Instrument," Orlando, FL: Proceedings of Institute of Nuclear Material Management, 2012.
10. Blanc, P.C., Menlove, H.O., Tobin, S.J., Croft, S., Favalli, A.; "An Integrated Delayed-Neutron, Differential-Die Away Instrument to Quantify Plutonium in Spent Nuclear Fuel," Journal of Nuclear Materials Management, vol. 40, no. 3, pp. 70-77, 2012.
11. Trellue, H.R., Galloway, J. D., Fischer, N. A. and Tobin S. J.; "Advances in Spent Fuel Libraries," Proceedings of Institute of Nuclear Materials Management conference, Palm Desert, California (July 14-18, 2013) and Los Alamos National Laboratory report LA-UR-13-24074.
12. Martinik, T., Henzl, V., Grape S., Jacobsson Svärd, S., Jansson, P, Swinhoe, M. T., Tobin, S. J.; "Simulation of Differential Die-Away Instruments Response to Active Interrogation of Asymmetrically Burned Spent Nuclear Fuel," Nuclear Instruments and Methods A, vol. 788, no. 11, July 2015.